Status of Research on Deposition of Superconducting Films for RF Accelerating Cavities

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Abstract – The paper describes status of research on the deposition of superconducting films for RF accelerating cavities. New UHV arc-based devices with planar or cylindrical niobium cathodes are presented. The main results and characteristics of arc-deposited thin superconducting niobium films, as well as the progress obtained recently in the formation of such films, are also described.

1. Introduction

Superconducting RF accelerators are mainly based on niobium bulk produced cavities. Copper cavities coated with thin niobium film would offer several advantages, such as better thermal stability, insensitivity to external magnetic fields, better mechanical stability, easier conditioning within the machine and connection to the cryostat, and finally lower cost. Since the late 80 s the magnetron sputtering technology has been applied for coating copper RF cavities with superconducting thin niobium films [1]. Unfortunately, despite past and recent efforts in the magnetron sputtering technology, a problem of the fast degradation of the quality factor for the coated cavities at higher fields has not been solved yet [2]. This Q degradation seems to be connected with chemical and physical properties of the niobium-copper system and probably with properties of the film-substrate interface. It is very known fact that during the magnetron sputtering deposition some amount of a working gas is incorporated into a growing film. The concentration of the commonly used Ar gas can reach 0.5%. The rare gas concentration in the film can influence several film properties.

In 2000 a new approach to the coating of cooper cavities was proposed – the vacuum arc deposition [3]. The vacuum arc has been known since the 70s to be a powerful technique to produce metallic and compound films with high density and very good adhesion. The arc technology promises some advantages, such as a high ionization ratio of metallic plasma, higher energy of ions in comparison with the magnetron sputtering technique and also a higher purity of the deposition process, due to the absence of a working gas. This usually results in the formation of a denser film and strongly reduces film defects, such as voids. The high ionization ratio of metallic particles emitted from the cathode make it possible to apply electric and magnetic fields for controlling the deposition process. The main disadvantage of an arc deposition is connected with the production of micro-droplets during the random movement of hot spots on the cathode surface.

In order to study the vacuum arc technique at ultra-high vacuum conditions for the production of high quality superconducting Nb-films, within a frame of the cooperation University of Rome "Tor Vergata" – IPJ-Swierk, under the Italian INFN research grant (ARCO project) financing and DESY support some efforts were undertaken in 2000. A number of UHV arc-based devices with planar and cylindrical cathodes have been constructed and investigated in the period from 2000 to 2003. So-far obtained results with thin Nb-films deposited upon sapphire and Cu substrates are very promising [4], [5].

Since 2004 our experimental program has been financed in the framework of the European Union "CARE" Program.

2. UHV Arc Based Apparatus

Details of our UHV arc-sources with a planar cathode have been described elsewhere [6], but some specific requirements and features of arc discharges at UHV conditions have to be reminded here for clearness.

The crucial role during the formation of a thin superconducting niobium-layer plays a cleanliness of the deposition process. For good superconducting film properties the partial pressures of water, nitrogen, oxygen, CO_2, hydro carbides, etc., must remain below $\approx 10^{-9}$ hPa during the deposition. The pumping system must, therefore, be totally oil-free and all parts of the deposition system must be designed and built in accordance with UHV technology requirements. In our case, all vacuum chamber components and accessories, as well as all vacuum connections, were fabricated using only high purity materials: stainless steel, OFHC copper and high quality ceramics shielded from the arc. The cathode and all parts accessible to the arc were made of ≥ 250 RRR Nb only. The base pressure of the order of 10^{-10} hPa is reached after one night baking of the whole system at 150 °C. To check the composition of the residual gases before and during the coating, the vacuum chambers are equipped with Residual Mass Analyzers (RGA).

In the first period of our investigations we based on the most widely used method of an arc initiation - high-voltage breakdown producing plasma by the evaporation of a thin metallic film along a dielectric surface. We applied 6 kV pulses to the thin niobium rod placed within a short Al_2O_3 tube. Recently, in the all our UHV arc-sources we use laser beams for triggering of arc discharges. Several triggering techniques have been tested from the point of view of operational reliability and cleanliness. We found that the best laser technique, using a laser beam focused on the cathode through a vacuum window, ignites the UHV arc without introducing any additional impurity whatsoever. Two different lasers have been successfully checked: a 60 mJ Nd:YAG laser with a 5-ns pulse width, operated with the repetition rate up to 20 Hz, has been used within the planar arc device in Rome, and a modified 700 mJ ruby laser, with 50 ns pulse width and relatively low repetition rate, has been applied within the linear arc device in Swierk. The laser ignition was decisive in improving the film superconducting properties.

At present four different UHV systems are in the operation at the University of Rome "Tor Vergata" and at IPJ in Swierk. Two devices are equipped with the planar arc sources. One of these planar arc sources is equipped with a 90° magnetic filter, to make possible a comparison of films produced with and without filtering. The picture of two UHV planar arc systems is shown in Fig. 1.

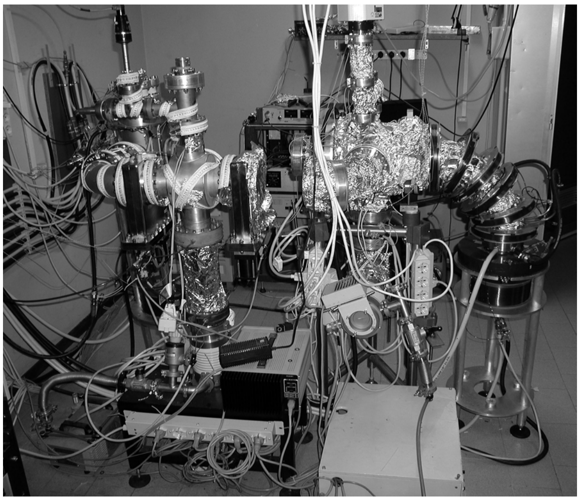

Fig. 1. Picture of two UHV planar-arc systems constructed at the "Tor Vegata" University in Rome

In future we will try to introduce metallic plasma by means of magnetic and electric fields, but the uniform coating of 1.5-GHz copper cavities (e.g. those of the TESLA type) by means of a filtered planar arc, still needs further R&D. An alternative being explored is that of using a linear geometry arc (cylindrical cathode) device that ideally meets the requirement of easily coupling to a single or multiple-cell cavity. The cylindrical cathode of such a system can in fact be placed along the cavity axis (like that of a cylindrical magnetron), with the arc discharge moving along it, either spontaneously or being magnetically driven.

The prototype linear arc source was designed and realized also in accordance with UHV technology requirements. The cathode (450 mm in length and 34 mm in diameter) was made from a RRR150 niobium tube, and it was directly water cooled. Niobium /OFHC-Cu/stainless-steel electron-welded transitions were used to prepare vacuum tight connections. For a displacement of the arc discharge along the cathode, a remote-moved permanent magnet was placed inside the niobium tube. Such a cylindrical arc source was installed on the axis of a copper cavity placed inside a large vacuum chamber (at oil-free HV conditions) to check possibility of the cavity coating.

The single-cell copper cavity of the TESLA type, which was coated with pure Nb, is presented in Fig. 2.

Fig. 2. TESLA type copper cavity, which was coated with a pure Nb layer at IPJ in Swierk

Under the real conditions the copper cavity should play a role of a vacuum chamber and of the anode simultaneously, and the whole system should of course guarantee the UHV conditions. Two such devices have been constructed at the "Tor Vergata" University in Rome and at the IPJ in Swierk. Recently they have been tested and put in the operation.

The schematic drawing of a new UHV set-up with the linear-arc source, as designed for single-cell cavity coating in the IPJ-Swierk, is shown in Fig. 3. A picture of this system is presented in Fig. 4.

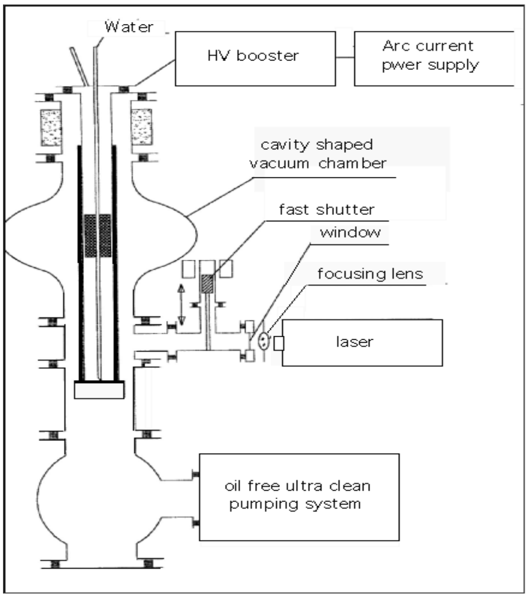

Fig. 3. Schematic drawing of the UHV set-up equipped with the linear-arc source

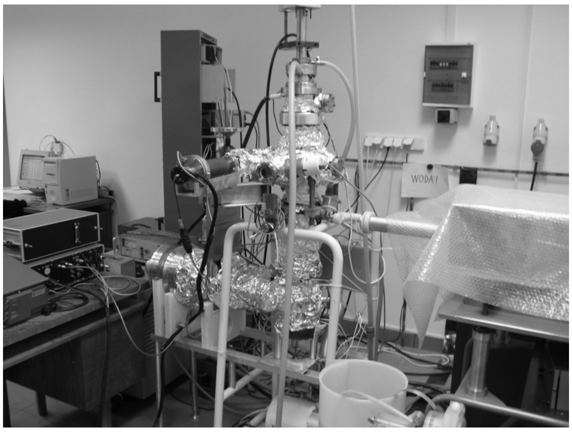

Fig. 4. Picture of the new UHV set-up constructed at the IPJ in Swierk

3. Formation and Properties of the Arc Deposited Niobium Superconducting Films

In order to coat sapphire and Cu substrates in the planar cathode system, they were mounted on a sample holder consisting of a massive Cu (OFHC) flange placed at a distance of about 50 cm from the cathode. The holder was kept at a constant temperature during the deposition process, and one could fasten up to 6 samples simultaneously. The sample holder was electrically insulated from the vacuum chamber and a bias of 20–100 V could be applied to the substrates. The lowest possible arc current for the stable operation in the present DC mode was found to be about 60 A, while the available cooling power did set a 140 A upper limit to the arc current. The deposition rate, achievable with the unfiltered planar arc source, can be very high. Its precise value depends on many factors, such as arc current, cathode material, geometry, applied fields, etc. In our planar system, operated with a current of 120–140 A, the deposition rate was ≈ 10 nm/s, while with the present not-optimized configuration of our filtered apparatus it was lower by a factor of ≈ 5.

The sample temperature during depositions was recorded by means of a set of thermocouples. Most of the samples were deposited at a room temperature, and only a few at higher (100–200 °C) temperatures. As above mentioned, the base pressure in our systems was in the 10^{-10} hPa range, it increased up to 10^{-6}–10^{-7} hPa as soon as the arc discharge started, and it remained almost stable throughout the deposition. The residual gas pressure during the discharge was found to be almost exclusively due to hydrogen. Its partial pressure was usually more than 3 orders of magnitude higher than that of other contaminants, as one can see in Fig. 5.

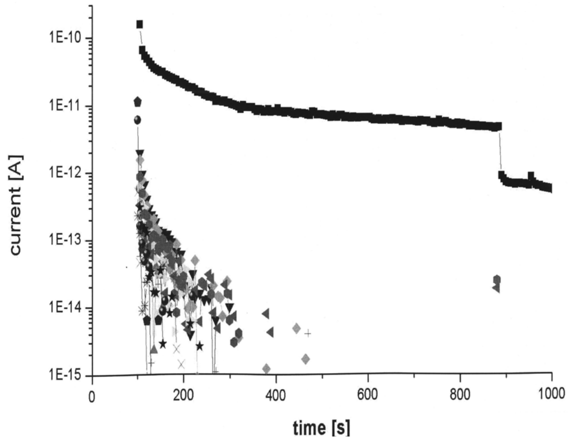

Fig. 5. Behavior of the partial pressure of different gases during the vacuum arc discharge

The UHV arc-deposited layers, mainly upon the sapphire substrates, have been characterized by measuring their Residual Resistivity Ratio (RRR) and the critical temperature (T_c). In fact the critical temperature of the deposited material is very sensitive to impurities, and even very small amounts of oxygen in the Nb-film can lower its T_c value significantly. The Nb Residual Resistivity Ratio (RRR defined as the resistivity at a room temperature divided by the resistivity at the temperature of 10 K) is also very sensitive to impurities; typical RRR values for Nb films deposited by sputtering at a room temperature range from 2 to 10, and films with RRR ≈ 25 are obtained either using Kr instead of Ar (as the auxiliary gas) or raising the substrate temperature to ≈ 250 °C.

The morphology of the deposited layers was also studied using the X-ray diffraction technique and the Atomic Force Microscope (AFM). The RRR of our 1.5-μm-thick Nb films, which were deposited upon sapphire substrates at a room temperature and under the typical UHV conditions described above, ranges from 20 to 50, and the typical averaged value was 40. A record value of RRR = 80 was obtained by heating the substrate to about 150 °C. The transition to superconducting state for this sample is shown in Fig. 5.

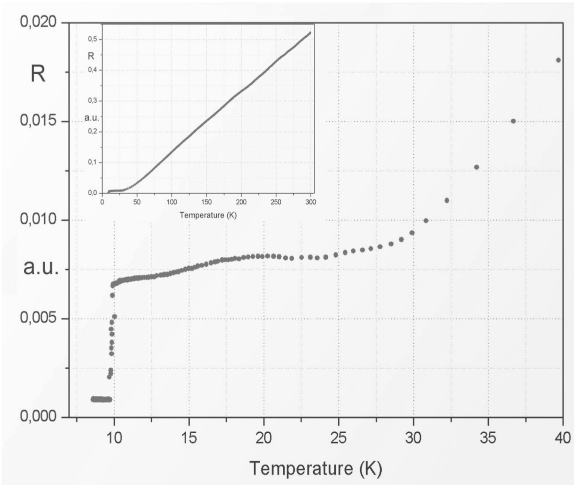

Fig. 5. Transition curve of the sample with RRR 80

The critical temperature (T_c) and critical current density (J_c) of the deposited films were measured using an inductive method. Typical results, as obtained for several samples, are shown in Fig. 6.

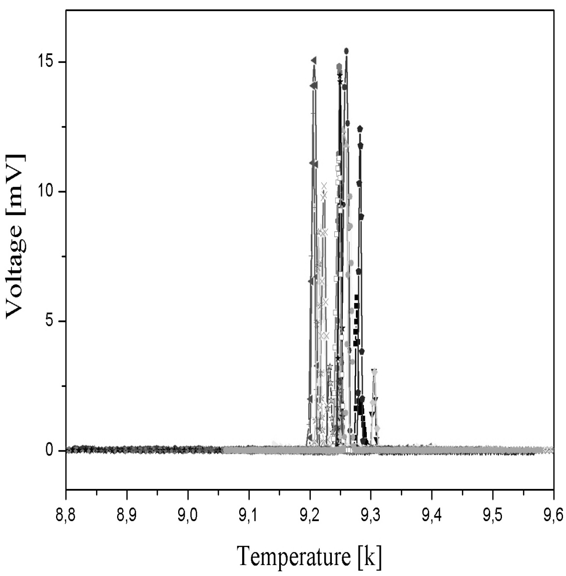

Fig. 6. Transition curves of a number of Niobium film samples, UHV arc deposited on copper and sapphire

Differences in T_c, as compared to the high purity bulk Nb value of 9.26 K, are small and can be in part attributed to small temperature differences between the thermometer and sample.

No dependence is observed on either film thickness, in the 1–2 μm range, or on arc current. This indicates that our films are less stressed than magnetron sputtered ones. That indication was confirmed by the XRD analysis (in a θ/2θ configuration) yielding a lattice parameter in the 0.3308–0.3318 nm range, i.e. a value close to the 0.3306 nm value for the bulk Nb. A further confirmation comes from the sharp transition widths (< 0.02 K), close to the ≈ 0.01 K value of the pure bulk Nb.

5. Micro-Droplets

The main inconvenience of the arc coating is the production of micro-droplets. In our case, micro-droplets composed of high purity molten Nb are not expected to contaminate the film, but they increase its surface roughness, and in a high electric field environment they may become field-emitters. The presence of micro-droplets upon the surface of our films was studied by means of optical- and electron-microscopy. Using a 500X magnification optical microscope, pictures of the sample surface were taken at 10 different randomly chosen locations and analyzed using a LabView computer code. It measured and recorded the number and size of the droplets present in the microscope (fixed area) observation field. The distribution of the droplet density as a function of the droplet size, as measured upon the surface of 4 different samples deposited under comparable conditions, is shown in Fig. 7.

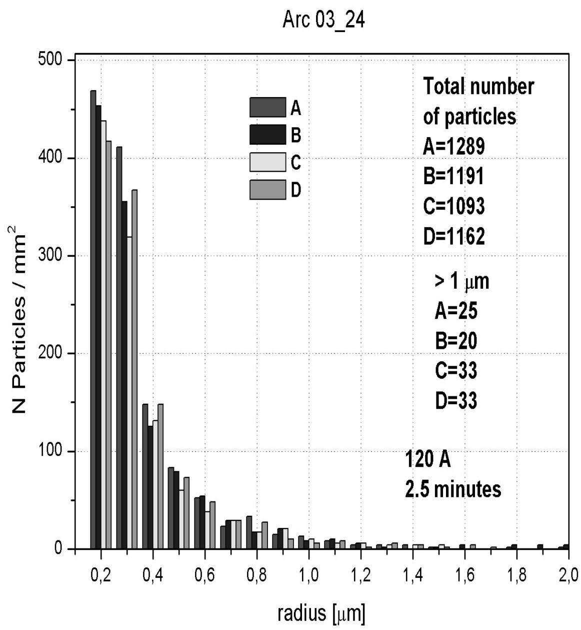

Fig. 7. Typical distribution of the droplet density, as measured upon surfaces of (A, B, C and D) samples coated under the same conditions

It should be noted that dimensions of the most observed droplets (200–300 nm) are comparable to size of the Nb-film grains whose boundaries can be seen in the picture presented in Fig. 8.

One can thus expect that many such droplets have become embedded in the growing film. Numerous larger droplets (> 1 μm) are also present, and they may become sources of the field emission from the coated surface of an RF cavity. Such a large droplet is shown in Fig. 9.

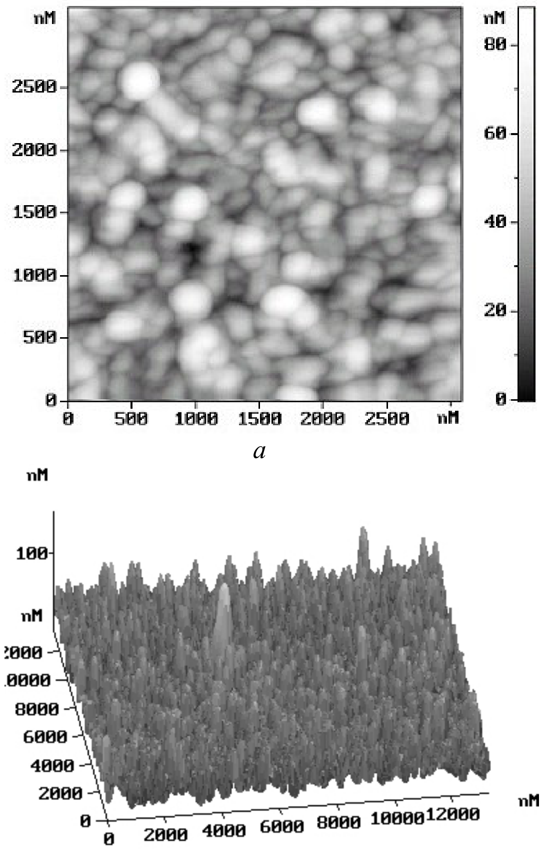

Fig. 8. AFM picture of a niobium film deposited on sapphire: *a* – Nb grains are visible (average dimension 200 nm); *b* – three-dimension view of the surface

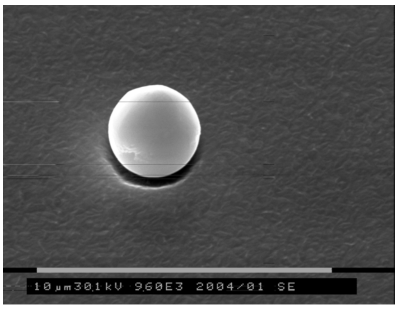

Fig. 9. SEM picture of a large Nb micro-droplet upon the film deposited on the sapphire substrate

Magnetic filtering can drastically reduce the number and dimensions of the micro-particles. Quantitative data on the reduction factor have not so far been obtained because (after filtering) the number of droplets in the observed field has been below the detection threshold.

6. Summary

Several cathodic-arc sources working in UHV conditions have been designed and constructed to study the deposition of super conducting Nb films. Results obtained with sample Nb films are promising, because the films with "bulk-Nb-like" properties were produced. Their RRR values and grain sizes are larger, as compared to Nb magnetron sputtered films deposited at the same temperature. They also appear to be less stressed and more randomly oriented. A filtered UHV arc system was also used to produce quasi micro-droplet-free samples. It has been shown also that by means of the linear arc it is possible to coat an inner surface of the 1.3-GHz copper cavity.

7. Acknowledgements

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