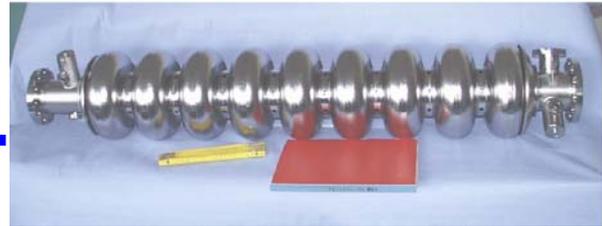




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### Thin superconducting niobium-coatings for RF accelerator cavities

J. Langner<sup>a</sup>, L. Catani<sup>b</sup>, A. Cianchi<sup>b</sup>, R. Mirowski<sup>a</sup>, J. Lorkiewicz<sup>b</sup>, D. Proch<sup>c</sup>, R. Russo<sup>b</sup>,  
M.J. Sadowski<sup>a</sup>, P. Strzyzewski<sup>a</sup>, S. Tazzari<sup>b</sup>, J. Witkowski<sup>a</sup>

<sup>a</sup>The Andrzej Soltan Institute for Nuclear Studies, Swierk, Poland

<sup>b</sup>University Tor Vergata, Via della Ricerca Scientifica 1, 001333 Roma, Italy

<sup>c</sup>DESY-MHF, Notkestr. 85, 22603 Hamburg, Germany

### Abstract

The paper describes efforts of four institutions which are engaged in the realization of the Work Package 4 (Thin Film Cavity Production) of the Joint Research Activity (JRA-1) within a frame of the Coordinated Accelerator Research in Europe (CARE) program. JRA-1 is aimed at developing superconducting RF technology, mainly methods for producing superconducting Nb coated copper cavities which might ensure higher accelerating fields, lower RF losses and considerable reduction of costs, as compared with the present state of art. WP4 is thus focused on the development of a new method to produce thin Nb-coatings by means of arc discharges performed under ultra-high vacuum (UHV) conditions.

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### ABSTRACT

The paper describes efforts of four institutions which are engaged in the realization of the Work Package 4 (Thin Film Cavity Production) of the Joint Research Activity (JRA-1) within a frame of the Coordinated Accelerator Research in Europe (CARE) program. JRA-1 is aimed at developing superconducting RF technology, mainly methods for producing superconducting Nb coated copper cavities which might ensure higher accelerating fields, lower RF losses and considerable reduction of costs, as compared with the present state of art. WP4 is thus focused on the development of a new method to produce thin Nb-coatings by means of arc discharges performed under ultra-high vacuum (UHV) conditions.

**Keywords:** vacuum arc discharge, UHV conditions, thin film deposition, superconductor

### 1. INTRODUCTION

At present days the superconducting RF-cavities with high electric fields for the acceleration of charged particles in high-energy accelerators are made of purified ( $RRR \geq 300$ ) niobium (Nb) or of copper (Cu) coated with a thin (a few  $\mu\text{m}$ ) Nb-film. The advantages of the latter technique, in a comparison with the former one, are mainly better thermal conductivity for the same mechanical strength and much lower sensitivity to external magnetic fields [1]. The largest scale application of this technique, that allowed also reduce the manufacturing costs significantly, has been the construction of the acceleration system for the LEP (Large Electron-Positron collider) at CERN [2]. It consists of several hundreds of 352-Mhz, four-cell Nb-coated copper (Nb/Cu) cavities operated at 4.2 K. The system have been operated reliably at field values up to 8 MV/m and quality factors (Q) of the order of  $4 \times 10^9$  for many thousands hours. It has proved, during the operation at relatively low frequency and at a LHe temperature, advantages of the described technique over the bulk Nb technology. On the other hand, at the much higher field levels, of the order of 20-40 MV/m, at Q values above  $10^{10}$ , which are required by today's very high-energy, high-power superconducting accelerators, the cavities must be operated at a super-fluid-He temperature and at frequencies higher than 1 GHz. In this parameter region, where the BCS resistance becomes negligible and resistive losses are dominated by the residual resistance depending on the peak electric field, the best performance is obtained with bulk-Nb cavities. It should be noted that the R&D effort by major laboratories and institutions in this field has been mainly focused on the bulk-Nb technology. This reason is that the latter technology, being much better known, held a promise of and actually led to the needed results faster. Nevertheless, due to the advantages of the Nb/Cu cavities there are undertaken efforts to reach the same performance as with the bulk-Nb structures. Therefore, unanimously recognized studies of the Nb/Cu alternative have been continued.

The goal of the CARE-JRA1-WP4 program described in this paper is to try and improve the performance of Nb-coated cavities. R&D are run by using a technology of the cathodic-arc deposition in order to form superconducting films different from those produced by the magnetron-sputtering developed at CERN to produce the LEP cavities [3]. The main institutions engaged in the described program are the IPJ in Swierk, Poland, and the INFN-Roma2 in Rome, Italy. The IPJ team is responsible for the WP4.1 task (Linear-Arc Cathode Coating), while the INFN-Roma2 team is

\*langner@ipj.gov.pl; phone +48 22 7180537

responsible for the WP4.2 task (Planar-Arc Cathode Coating). These two tasks are based on different electrode geometries, what in the principle offers somewhat different advantages, but the both explore the same principle and are carried out in the close scientific collaboration between the two teams. A support is also acknowledged from the “Tor Vergata” University of Rome (Italy), INFN-LNF in Frascati (Italy), CERN in Geneva (Switzerland), Cornell University in Ithaca (USA), DESY in Hamburg (Germany), Istituto di Cibernetica “E. Caianiello” at CNR Pozzuoli (Italy), Università di Napoli “Federico II” in Napoli (Italy), HCEI in Tomsk (Russia) and INFN-in Napoli (Italy).

## 2. PERFORMANCE OF COATED CAVITIES

The main advantage of the Nb/Cu cavities, besides possible reduction of the fabrication costs, is to solve the problem of the heat dissipation, which is induced by the low heat conductivity of pure Nb. A thin Nb film determines the cavity superconducting behavior, while the high heat-conductivity copper substrate, thick enough to provide mechanical stability, does effectively transfer the heat produced inside the cavity to the liquid-He bath. It significantly increases the tolerable heat production level. In addition, the film magnetic field pinning energy is very high (of the order of  $10^9$  N/m<sup>3</sup>), so that external magnetic fields of the order of the Earth-field do not create free fluxons leading to the energy dissipation [4]. It has also been shown [5] that superconducting properties of the film such as  $H_{c1}$ ,  $R_{BCS}$ ,  $T_c$  and  $R_{fl}$ , depend on the film purity only, and therefore on the mean free path  $\lambda$ , so that that the purer the film the closer are its parameters to those of the bulk metal.

In the light of such observations the present magnetron sputtering techniques appears to have some drawbacks. First of all, the magnetron-sputtered cavities, coated in an atmosphere of inert gas (typically Ar), are inevitably contaminated by the auxiliary gas itself. In addition, the observed film porosity, caused not only by the substrate roughness but also by angles of incidence of the Nb atoms on the cavity walls, added to the low-energy of the metal ions - may possibly be an important factor limiting the cavities performance. Both drawbacks are (if not eliminated) expected to be strongly reduced by depositing the Nb film by means of the UHV cathodic-arc technique. This method of the arc coating under ultra-high vacuum (UHV) conditions promises superior properties of the deposited superconducting films. The UHV arc technique, as briefly discussed below, is also amenable to the deposition of superconducting composite materials with higher  $T_c$  than that of pure Nb, such as NbN.

## 3. VACUUM ARC

The name “vacuum arc” (VA) is usually applied to a low-voltage (20-40 V), high-current discharge (50-200 A) initiated within high vacuum (HV:  $10^{-5}$ - $10^{-6}$  hPa) and taking place in vapors of the cathode material. The discharge develops on the cathode surface in microscopic hot spots with a current density within the range of  $10^4$ - $10^6$  A/cm<sup>2</sup>. Such spots can move randomly over the cathode surface with velocities of  $10^2$ - $10^4$  cm/s. Evaporation products, whose phase composition is determined primarily by the cathode material, form plasma channel of a density equal to  $10^{11}$ - $10^{12}$  cm<sup>-3</sup>, at the significant concentration of multiply charged ions which can be collected and guided to the anode by properly applied magnetic fields. The cathode mass loss is generally expressed in micrograms per coulomb and the generated products contain vapor, microscopic droplets and variously charged ionized particles. For refractory metal arc-plasmas the fraction of ionized atoms may be as high as 90-100%. The facts that the ion energy is relatively high, in the range of 10-100 eV, as compared to 5-10 eV observed in the magnetron-sputtering technique, and the ions have high average charge, are the main features of the VA process. Because of a high degree of the ionization, energies of the ions may be further increased by applying a negative bias to a conducting substrate.

Such ions thus have sufficient energy to clean contaminant atoms off the surface prior to forming the coating, and sufficient mobility to diffuse to low free energy sites. High ion energy is thus the main factor responsible for much compacter films, with much stronger adhesion to the substrate than obtainable by other methods, as confirmed by molecular dynamics calculations. Therefore, the vacuum arc deposition offers an excellent approach to producing, at very high rates, pure metal-, alloy- and compound-films with excellent adhesion and density [6].

The main disadvantage to be overcome in the VA deposition is, as above mentioned, the ejection of micrometer-sized droplets (called often micro-droplets or macro-particles) from the cathode surface. Such micro-droplets are mainly ejected in directions close to the cathode surface plane with velocities from tens to hundreds m/s. Their production, inherently connected to the existence of high-power density hot-spots upon the cathode surface, depends on the cathode material and temperature as well as on discharge parameters, such as the arc current and applied magnetic field. The

dimensions of the micro-droplets (macro-particles) are usually within the range of 0.1-10  $\mu\text{m}$ . In the case of refractory metals the micro-droplet phase can amount to about 1% of the total evaporated material.

#### 4. ARC IN ULTRA-HIGH VACUUM CONDITIONS

The crucial role during the formation of a thin superconducting niobium-layer is played by cleanliness of the deposition process. In order to achieve good properties of the superconducting film, the partial pressures of water, nitrogen, oxygen,  $\text{CO}_2$ , hydro-carbides etc., must remain below  $\approx 10^{-9}$  hPa during the deposition process. The pumping system must, therefore, be totally oil-free and all parts of the deposition system must be designed and built in accordance with the UHV-technology requirements. In our case, all vacuum chamber components and accessories, as well as all vacuum connections, are 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 are made of pure Nb with  $\text{RRR} \geq 250$  only. The basic pressure of the order of  $10^{-10}$  hPa is reached after one night baking of the whole system at a temperature of 150  $^\circ\text{C}$ . The composition of residual gases before and during coating is monitored with a Residual Gas Analyzers (RGA). Typical examples are shown in Fig.1.

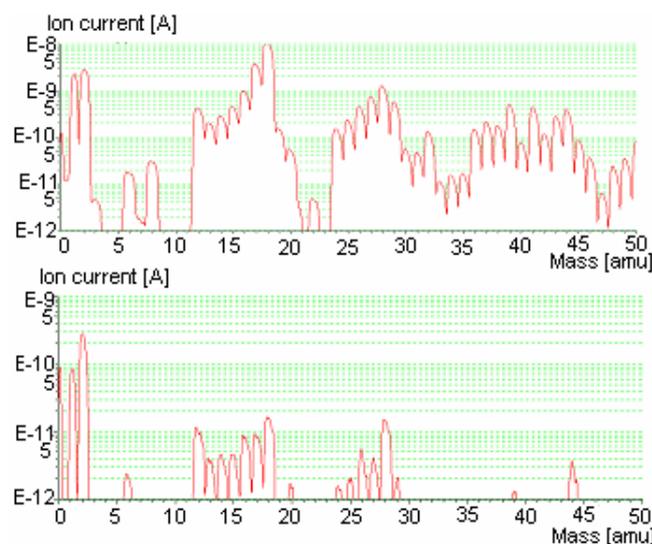


Figure 1: Typical RGA mass-spectra recorded before and after 24-hour baking at 150  $^\circ\text{C}$ .

The presented mass-spectra demonstrate very well differences between HV and UHV conditions, which are of primary importance for the deposition of pure films. Another question is the initiation of vacuum arc discharges.

The reliable triggering (ignition) of the arc discharges is often a problem, even in industrial arc-based devices. In HV systems, thin layers of gasses and impurities formed upon the surface of electrodes, are beneficial in that sense that they facilitate the starting of an arc discharge. Under UHV conditions the high-temperature baking of the vacuum chamber removes almost totally such layers. These effects, as well as requirements that all other sources of impurities must be effectively removed, make the arc ignition more difficult.

After testing many known triggering methods from the point of view of operational reliability and cleanliness, we have finally decided to use a laser beam focused upon the cathode through a vacuum-tight glass window. Then the arc is triggered extremely reliably without introducing any additional impurity whatsoever [7]. Two different lasers have been successfully tested: for the planar arc in Rome we used a 60-mJ Nd:YAG laser generating 5-ns pulses with the repetition rate up to 20 Hz, and for the linear arc in Swierk we applied a modernized 700-mJ ruby laser generating 50-ns pulses with a relatively low repetition rate. Recently, the both laboratories have used 100-mJ Nd:YAG lasers, and the mastering of the laser ignition technique was decisive for improving properties of superconducting films.

### 5. UHV ARC-BASED DEVICES

The UHV or HV arc deposition systems, which are in the operation in the Physics Department of the Tor Vergata University in Rome, and in the Department of Plasma Physics and Technology of the IPJ in Swierk, are listed in Table 1.

Device	Cathode geometry	Filtering	Vacuum	Film	Lab	Task
PAUHV-2	Planar	-	UHV	Nb, W	Rome	sample deposition
FPAUHV-1	Planar	+	UHV	Nb	Rome	sample deposition
CCLAUHV-2	Linear	-	UHV	Nb	Swierk	Cu cavity cell coating
CCPAUHV-1	Planar	-	UHV	Nb	Rome	Cu cavity cell coating
TSFPAUHV	Planar	+	UHV	Nb, Pb, Mg	Swierk	droplet filtering tests
TSFLAHV	Linear	+	HV	Nb	Swierk	droplet filtering tests
CPAUHV	Planar	-	UHV+N <sub>2</sub>	NbN, TiNbN	Rome	sample deposition

Table 1. Vacuum arc-based devices investigated in Rome and Swierk.

Five facilities are equipped with planar-arc (PA) sources. A scheme of the PA source with a magnetic filter is shown in Fig.2.

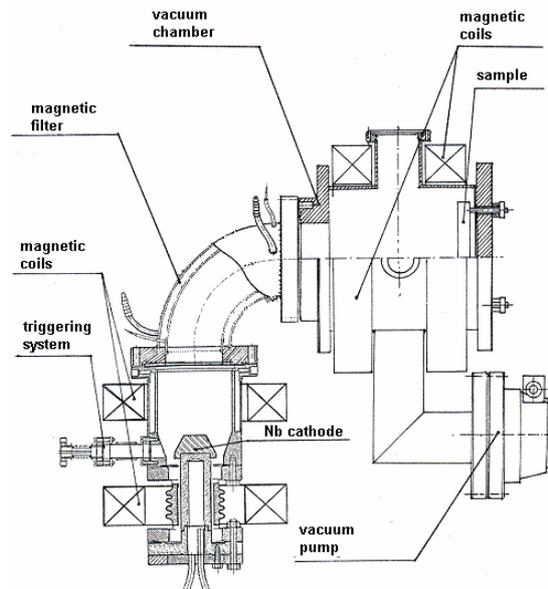


Figure 2: Schematic drawing of a typical planar-arc (PA) source with the magnetic filter used for the elimination of micro-droplets emitted from the cathode surface.

Three investigated systems (FPA) have been equipped with knee-shaped magnetic filters in order to enable a comparison of the deposited films (produced with and without filtering) and to study different filtering configurations. Two of these systems are shown in Fig. 3.

### 6. FORMATION AND PROPERTIES OF UHV ARC-DEPOSITED NIOBIUM SUPERCONDUCTING FILMS

In the PA systems there can be coated up to six sapphire- and Cu-substrates simultaneously. They can be mounted upon a sample holder consisting of a massive Cu (OFHC) flange, placed at a distance of about 50 cm from the cathode

and kept at a constant temperature during the whole deposition process. The sample holder is electrically insulated from walls of the vacuum chamber, so that a bias of 20-100 V can be applied to the coated substrates. The lowest possible arc current for the stable operation in the present DC mode has been found to be about 60 A, while the available cooling system of the anode has an upper limit of the arc current equal to about 140 A. The deposition rate achievable with the unfiltered PA source can be very high and its value depends on many factors, such as the arc current intensity, the cathode material, geometry, applied fields, etc. In our unfiltered system operated with arc currents of 120-140 A the deposition rate is 10 nm/s, while within the present not-optimized FPA configuration it is about 5 times lower.

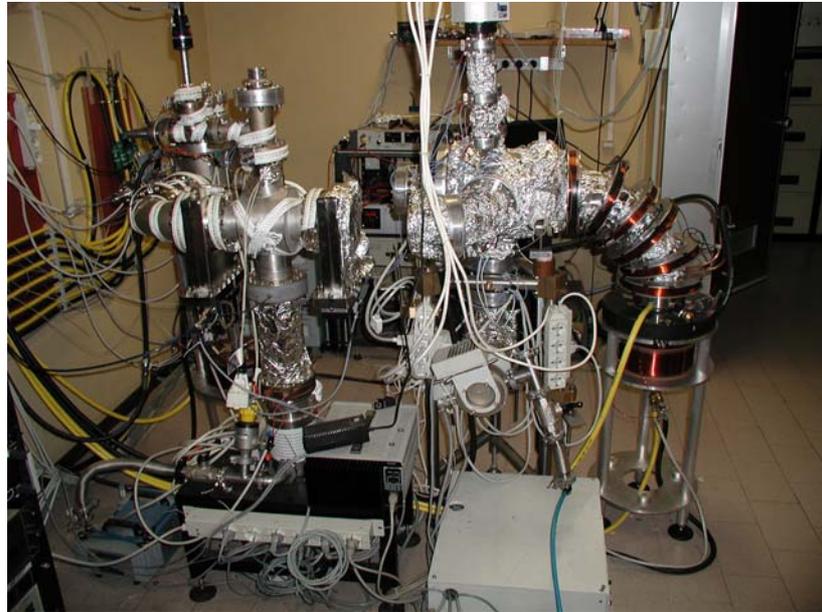


Figure 3: Two experimental systems equipped with planar-arc sources and magnetic filters, which are investigated in Rome.

The sample temperature during depositions is usually recorded by means of thermocouples. Most samples have so far been deposited at temperatures close to the room temperature, and only a few samples have been investigated at higher temperatures (100-200 °C). As above mentioned, the residual pressure in our systems is usually set within the  $10^{-10}$  hPa range. The pressure increases up to  $10^{-6}$  -  $10^{-7}$  hPa when the arc discharge starts, and it remains almost stable at the latter value throughout the deposition process. The gas pressure rise during the arc discharge is found to be almost exclusively caused by hydrogen, which partial pressure is usually more than 3 orders of magnitude higher than that of other contaminants, as shown in Fig.4.

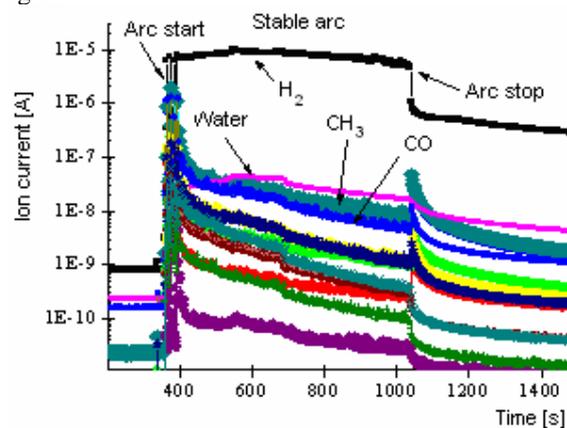


Figure 4: Behavior of the partial pressure (in ion-current units) of residual gases during the arc discharge. The downward slope is due to the pumping effect of a freshly deposited film.

The UHV arc-deposited Nb-layers on sapphire substrates have been characterized by measuring their critical temperature  $T_c$  and Residual Resistivity Ratio (RRR: defined as the resistivity at room temperature divided by the resistivity at 10 K). The critical temperature of the deposited material is in general very sensitive to impurities, e.g. very small amounts of oxygen in the Nb-film can lower its  $T_c$  value significantly. The Nb RRR 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 the Nb-films with  $RRR \approx 25$  are obtained either using as the auxiliary gas Kr instead of Ar or raising the substrate temperature to  $\approx 250$  °C.

The RRR values of our 1.5- $\mu\text{m}$ -thick Nb films, which were deposited upon the sapphire substrates at a room temperature, under the typical UHV conditions described above, range from 20 up to 50. A record value of  $RRR = 80$  was obtained by heating up the substrate to the temperature of 150° C [8].

The critical temperature ( $T_c$ ) and critical current density ( $J_c$ ) values of the deposited Nb films were measured by means of an inductive method. The best samples have shown the values identical to bulk Nb, i.e.  $T_c = 9.26$  K,  $\Delta T_c = 0.02$  K and  $J_c = 3 \times 10^7$  A/cm<sup>2</sup> [9,10]. Typical results, as obtained for several samples, are shown in Fig.5.

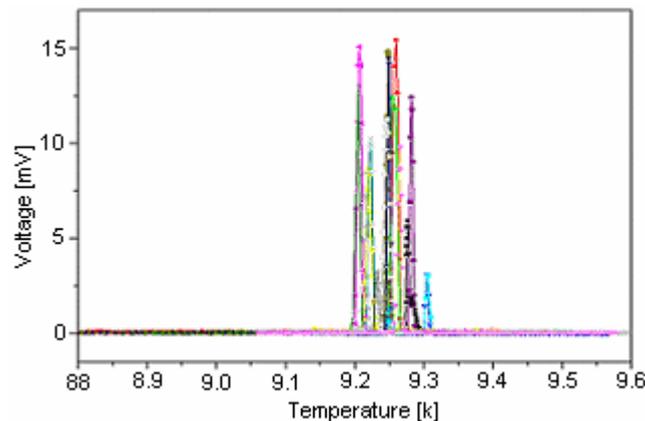


Figure 5: Transition curves measured for several samples of Nb-layers which were deposited upon Cu- and sapphire-substrates by means of UHV arc discharges.

The observed differences in  $T_c$  value, as compared to that of the high-purity bulk Nb value equal to 9.26 K, are small and can partially be attributed to small temperature differences between the thermometer and the investigated sample.

## 7. Macro-particle filtering

The main disadvantage of the arc coating is the production of micro-droplets (macro-particles). In our case, the micro-droplets of high-purity molten Nb are not expected to contaminate the film, but they can increase its surface roughness and, and within a high electric field environment they may become field-emitters. Their presence upon the coated surface of our films was studied by means of the optical- and electron-microscopy. Using an optical microscope with a 500X magnification, there were taken pictures of the sample surface at 10 different points chosen randomly. Those pictures have been analyzed using a LabView computer code, which enabled us to measure and record the number and sizes of the micro-droplets visible in the microscope viewing field (fixed area). The surface density of the recorded micro-droplets as a function of their sizes, as measured upon 4 different samples deposited under almost identical experimental conditions, is shown in Fig.6.

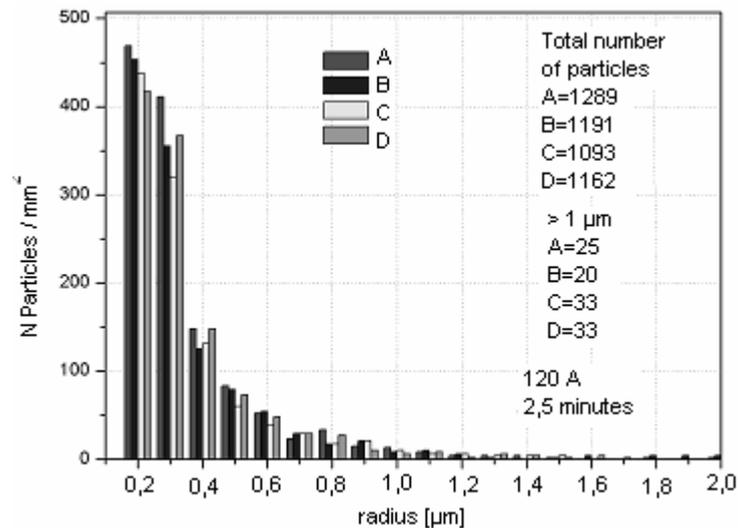


Figure 6: Typical histogram showing the surface density of the micro-droplets visible upon surfaces of four UHV arc-deposited samples (denoted by A, B, C and D).

The dimensions of most observed droplets are comparable in size (200-300 nm) to the Nb film grains [9]. One could expect that such micro-droplets become embedded in the growing film. Nevertheless, since such macro-particles can increase the surface roughness and possibly porosity as well as the field emission capacity, for our future applications (within high-field RF cavities) they must be eliminated by proper filtering. This can be done by magnetic deflecting the plasma flow, so that the coated substrate could not see the cathode surface directly. The first magnetic filters for the elimination of the micro-droplets from HV-arc systems were developed by Aksienov many years ago [11], and the main principle of their operation has remained the same. Various filtering systems have intensively been developed in different laboratories especially during the last 2 decades, and one can observe in large progress on this field [12]. Unfortunately, the construction of magnetic filters for the UHV conditions is more difficult than that for of the filters operated at higher basic pressures. The main differences are higher requirements as regards the constructional materials and their vacuum behavior, as well as necessity of the application of additional heaters for baking. Moreover, the magnetic filters for their long and stable operation must be cooled by an appropriate water flow.

In the magnetic filter, which has been designed at IPJ and is being applied in the both collaborating laboratories, the combination of heating and cooling systems was realized by the use of two coaxial metal walls. The inner wall is heated only during the baking process and it is cooled down during the arc operation. The both walls were made of a non-magnetic stainless steel. Both ends of the walls were closed by CF100 flange. Additionally, in order to control temperature changes, a thermo-couple was placed upon the inner wall of the filter. The constructional details of the described magnetic filter, as designed for the UHV arc system, are shown in Fig.7.

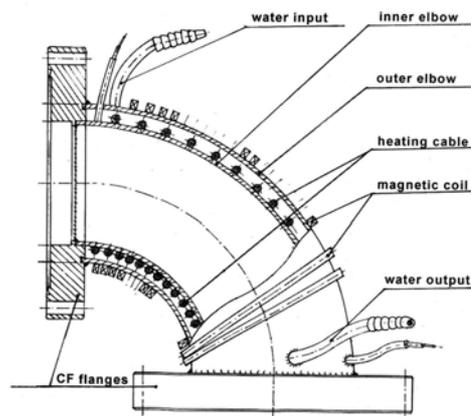


Figure 7: Design drawing of the UHV magnetic filter.

The distribution of magnetic field lines within the filter and near cathode region, as calculated by means of a Maxwell 2D-program, is shown in Fig.8.

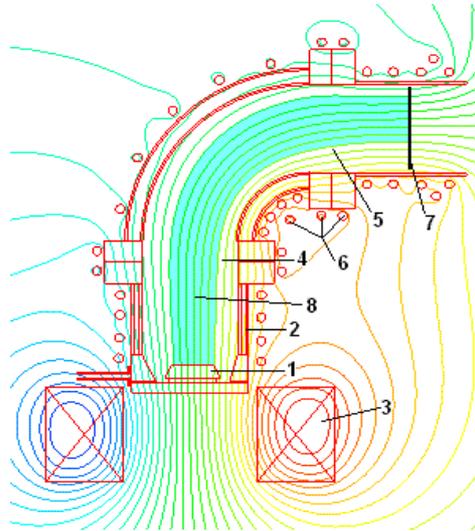


Figure 8: Distribution of magnetic field lines in the applied planar-arc source and magnetic filter channel. 1 – cathode, 2 – anode, 3 – focusing coil, 4 – filter inlet, 5 – filter exit, 6 – high-current cable, 7 – ion collector position, 8 – plasma stream.

When the magnetic filter is applied, the deposition rate is reduced by a factor of 5 with respect to that of the unfiltered system. The magnetic filter can reduce the number and dimensions of the deposited micro-droplets (macro-particles) drastically, as one can observe in Figs. 9 and 10.

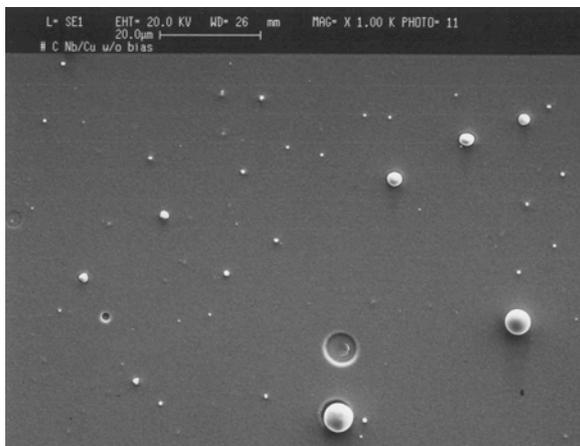


Figure 9: Surface of the Nb coating obtained without magnetic filtering.

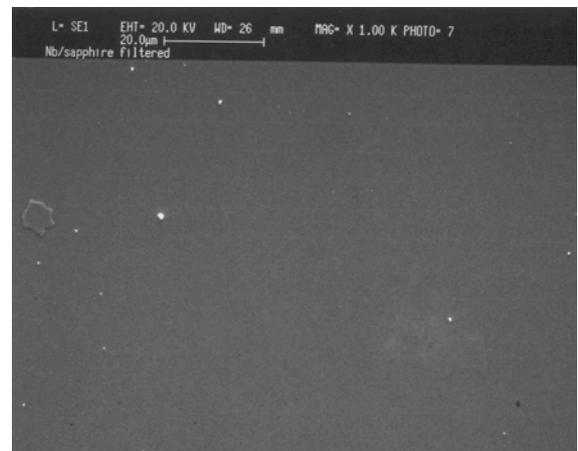


Figure 10: Surface of the Nb coating obtained with the use of the filtered UHV arc.

### 8. CAVITY COATING

The experimental results obtained so far with the deposition of thin superconducting Nb films upon the sapphire- and Cu-substrates are very promising, but it is obvious that that the coating of the whole inner surface of the copper cavity needed for the operation at 1.5 GHz frequency (relatively small dimensions), is not a simple task.

For the coating of inner surfaces of such accelerating structures, there were proposed various approaches. First one has been based on a linear geometry of the arc source. A cylindrical cathode of the linear-arc can be placed along the cavity axis, similar to the system with the cylindrical magnetron used in CERN. Changing a position of a permanent magnet, which might be located inside of the tubular Nb cathode, one can drive the arc discharge along the symmetry axis in order to obtain uniform coating of a single cavity or multi-cell structure. A scheme of such a system, which has been designed at IPJ, is shown in Fig.11.

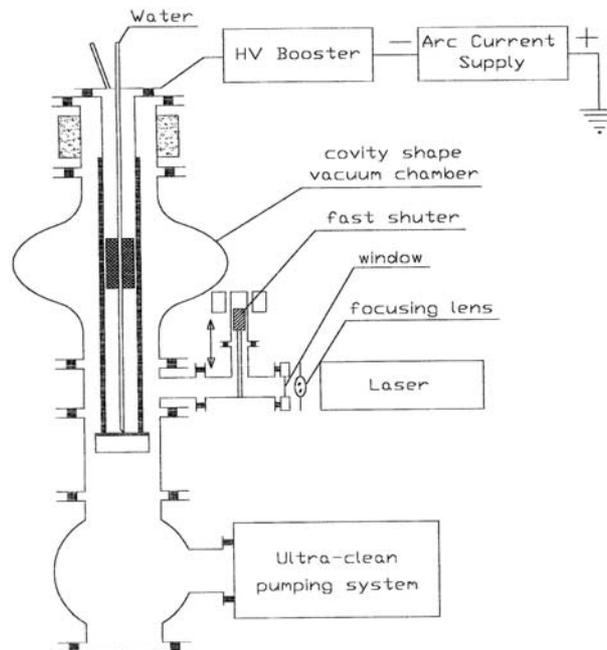


Figure 10: Schematic drawing of an UHV apparatus with the linear-arc source for single cell cavity coating.

One can expect that in the described configuration the main problem will be connected with the design and operation of an appropriate cylindrical filter for the elimination of micro-droplets. Research on this problem has just been started at IPJ in Swierk. In order to verify the principle of the linear (cylindrical) arc, a new UHV-apparatus equipped with the linear-arc source for coating single cavities was put into operation in Swierk in February 2005 (see Fig.12).



Figure 12: UHV system with a linear-arc and a test bed of the knee-shaped magnetic filter, which is operated in Swierk.

In future we would like to apply UHV arc-produced Nb-plasma filtered by means of magnetic and electric fields. A new technique based on coating inner surfaces of RF-cavities with two planar-arc sources, which might be placed at the both ends of the single cavity (or a multi-cell) structure, with the use of two external magnetic filters, is also under development.

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