ENERGETIC CONDENSATION GROWTH OF Nb THIN-FILMS FOR SRF APPLICATIONS

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Abstract

AASC, Jefferson Lab and NSU conduct research into new SRF thin-film coatings by first characterizing the materials properties such as morphology, grain size, crystalline structure, defects, and impurities, then measuring properties such as T_c and RRR and following this with 'in-cavity' RF measurements of the Surface Impedance of the films at cryogenic temperatures. These progressive steps are essential to the eventual design of SRF accelerator structures and to measure Q-slope and other performance parameters at high fields. We have recently produced Nb superconducting thin-films with crystal grain sizes in the range of 50µm using our proprietary CED^{TM} cathodic arc technique. RRR of ~129 at T_c of 9.2K was measured in a film grown on a-plane sapphire heated to 400^{o}C. At 20^{o}C, the RRR dropped to ~4. Energetic condensation using cathodic arcs produces non-equilibrium fast ions (~50-100eV). These ion energies are much higher than typical sputtering energies. When such energetic condensation is complemented by substrate biasing (to ~200-300eV) the incident ion energy is further increased, allowing growth modes that would otherwise require much higher substrate temperatures. Data are presented for pure Nb films using SEM, EBSD, XRD and a Surface Impedance Characterization RF cavity.

INTRODUCTION

Most RF particle accelerators worldwide utilize RF cavities made from a conventional conductor such as copper to accelerate the particle beam. A few large research accelerators such as the Continuous Electron Beam Accelerator Facility (CEBAF) at Thomas Jefferson National Accelerator Facility (Jefferson Lab, or JLab) and the Spallation Neutron Source (SNS) use superconducting radio frequency (SRF) accelerating cavities. SRF cavities consume less power than conventional cavities to produce a given accelerating gradient, even when the additional energy cost of the cryogenics system is taken into account [1, 2]. However the cryogenics system costs do represent a significant portion of the operating expense of SRF accelerators. Operation at higher temperature would naturally reduce those costs. Niobium, the only presently accepted superconductor for SRF accelerators, has a transition temperature T_c of 9.3K, but a practical operating temperature of 2K at the typically used 1.3–1.5 GHz RF frequencies. Operation below T_c is required to minimize surface resistance and maximize critical magnetic field. The cavities are typically made of niobium metal which becomes superconducting when cooled to a few degrees above absolute zero. Increasing the operating temperature of the accelerator from 2K to 4.5K could cut the cryogenics system costs in half [1, 2] but would require use of an alternative superconductor with an operating temperature of 10K or higher. Since the RF (London) penetration depth on the cavity surface is only ~30nm, it is of interest to develop thin film coatings on the nano-scale (~100nm) for particle accelerators. One approach to reducing SRF accelerator costs is to replace the expensive, bulk Nb (~$300/lb) cavities with a Nb thin film deposited on a less expensive material, such as Cu (~$3/lb) or better yet, Al (~$1/lb). The added advantage of Al [3] over Cu is that the cavity can be cast instead of being machined, which is more expensive.

Pioneering work [4-6] on Cu cavities coated with Nb thin film has been done at the European Organization for Nuclear Research, known as CERN. By 1998, 272 copper 352 MHz cavities, Nb thin film coated via magnetron sputtering, were deployed for the Large Electron-Positron Collider (LEP) project. The circular LEP collider, with a circumference of 27 kilometers, was one of the largest particle accelerators ever constructed and has recently been replaced by the Large Hadron Collider (LHC). Future accelerator facilities, such as the proposed International Linear Collider (ILC), require high accelerating field ~35 MV/m and $Q \geq 10^{10}$. It was reported [6, 7] that at 1.7K the Nb thin film cavities for LEP had Q-drop to below 10^{10} at ≈15MV/m and to below 5×10^9 at ≈20 MV/m. Thus there is a motivation to better understand these limits and to improve Nb thin films' performance for future SRF accelerator cavities.

The payoff of higher temperature SRF cavities and cavities made out of thin-film coated Cu or Al has motivated a multi-year research program at Alameda Applied Sciences Corporation (AASC). The goal is to utilize energetic condensation to produce Nb-on-Cu films of sufficient quality to determine if the thin film RF properties are adequate for high power SRF applications such as particle accelerators. Thorough characterization of the surface morphology and RF properties of Nb-on-Cu is a necessary first step toward qualifying Nb coated Cu SRF cavities. Given the high-cost (>$100M) and infrequent occurrence of large SRF accelerator upgrades and construction, it is difficult to imagine acceptance of an alternative superconductor in SRF accelerators without demonstrated performance equivalent to (or better than) that of bulk Nb (2K). Implementation of Nb-on-Cu coated

cavities into commercial accelerators, our long-term goal, is likely to follow reliable operation on large accelerators.

EXPERIMENTAL CONFIGURATION

The experimental configuration for the CED™ chamber is shown in Fig. 1. The arc is formed between an on-axis cathode (1-cm diameter and 60-cm long) and a coaxial mesh anode (45-cm in length) at 1-cm radius. The arc is sustained by a PowerTen, Inc. power supply (100V, 200A). The repetition rate of the arc is 0.5Hz, limited at present by SCR switches, but may be increased by using IGBT switches to more rapidly turn off the arc between pulses.

The arc is triggered by a proprietary trigger system. The substrate is placed outside the anode. The entire assembly is enclosed in a 25-cm dia. CF vacuum spool. The vacuum spool is double-walled with a solenoid wound on the outside. The solenoid is capable of producing a peak magnetic field of 10mT in either the z^+ or z^- direction. The arc current is controlled by adjusting a ballast resistor in series with the cathode. Fig. 2 shows a photograph of the CED™ apparatus with a 2.2 GHz copper RF cavity adjacent to it.

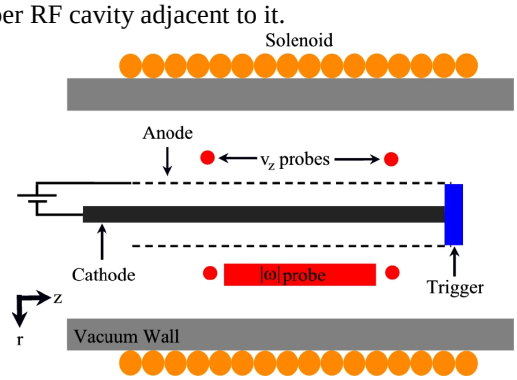

Figure 1: Illustration of the CED™ system.

Figure 2: Photograph of the CED™ system, showing a 2.2 GHz copper RF cavity for scale.

AASC's CED™ energetic plasma condensation deposition apparatus allows deposition of superconducting Nb thin films on Cu substrates heated to temperatures in the 300-450C range. This system is equipped with a substrate heater and temperature control system that is capable of operation up to 1200 ºC. The key process variables are substrate temperature, degree of annealing, substrate bias, deposition rate, and base pressure. Base pressures of ~$5x10^{-9}$ Torr are possible within this chamber and are measured and monitored using a SRS200 residual gas analyzer (RGA). In addition to substrate temperature control, the substrate can also be biased.

Energetic condensation occurs from a high-current arc (>10A) between the source material (cathode) and an anode, typically a robust refractory material, SS304 or OFHC Cu. The cathode material is ionized by the arc in localized regions of high current density (>10^5A/cm^2) called cathode spots. Fully ionized plasma and droplets of cathode material, commonly referred to as macro-particles, form a highly energetic plume. The macro-particles must be filtered from the plasma, but several filtering techniques are mature and commonly available [8]. Energetic condensation methods are known to produce compounds at high deposition rates (~50 Å/s) and reduced substrate temperatures compared to lower energy deposition methods such as magnetron sputtering, due to the population of non-equilibrium fast ions in cathodic arc plasmas. [8] It is important to control film microstructure at the grain level. Even if the films deposited are highly pure with proper stoichiometry, the grain size and grain connectivity are important. Deposition techniques with low ad-atom energy produce amorphous films and typically, high substrate temperatures and ex-situ anneal steps are required to form the large, highly connected grains necessary for high superconductivity. These techniques are incompatible with a Cu substrate as Cu cavities tend to get distorted if one uses temperatures that are higher than 400^0C, if the cavity is under vacuum. Therefore, annealing steps must be done at still lower temperatures. The CED™ energetic condensation process deposits from high-energy, non-equilibrium plasmas, enabling control of ion energy ***and*** relatively low (<400C) substrate temperature to tailor the film structure. A DC supply is available to bias heated substrates to ±1500V. Growth modes are realized that would otherwise require higher substrate temperatures with no bias. For example, with the substrate biased to ~1-2kV, sapphire can be grown at a temperature that is 200°C below that required using energetic condensation alone. [9]

When energetic condensation is augmented by pulsed substrate biasing (to several kV in (~1-10μs pulses)), incident ion energies are further increased, allowing further tailoring of film structure.

Deposition rates for cathodic arc process are high, even considering the inherent transmission efficiency of a macro-particle filter. Deposition rates of 5-20 nm/s are commonly reported. [10] The deposition rate is easily controlled since the mass liberated from the cathode is directly related to the total charge transferred by the arc. The charge transfer increases by increasing the magnitude and/or the duration of the current.

Nb DEPOSITIONS

Nb thin-film depositions were carried out in the CED™ apparatus on a-plane sapphire and Cu substrates. The sapphire coatings allowed measurements of RRR to be made, whilst the Cu coatings were used to study the influence of substrate temperature on film quality. Several tools were used to characterize the films: SEM, XRD and EBSD. In addition, RRR was measured by measuring the resistivity vs. temperature.

PROPERTIES OF Nb THIN-FILMS

Superconductivity in CED™ Deposited Films

Figures 3 and 4 show RRR measurements made on sapphire coated films that were deposited at 300°C (sample AASC-126-015) and 400°C (sample AASC-126-022) .

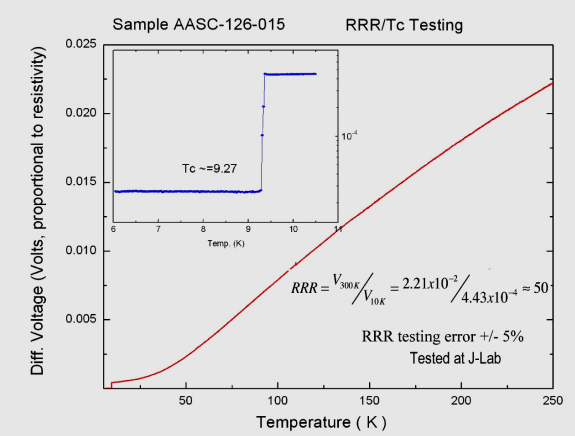

Figure 3: Superconducting transition (inset) and RRR in Nb thin-film coated on a-plane sapphire at 300°C.

The inset in Figure 3 shows the superconducting transition at 9.3K. The RRR (defined as the ratio of resistivity at 300K to that at 10K, is ≈50 in this case. A coating applied at room temperature showed RRR≈5.

Figure 4 shows the same data but for a coating that was applied at 400°C.

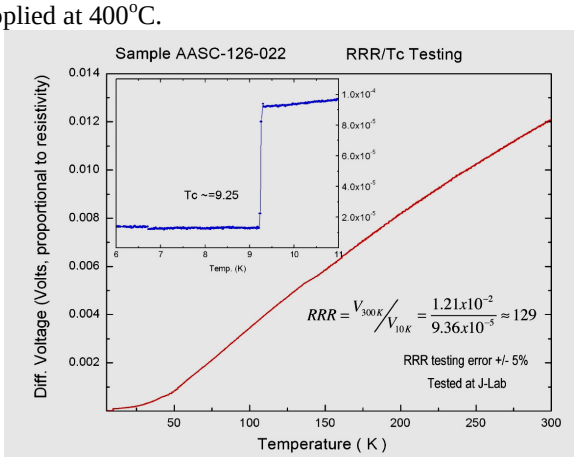

Figure 4: Superconducting transition (inset) and RRR in Nb thin-film coated on a-plane sapphire at 400°C.

Whereas the transition is not that much sharper in this case than at 300°C, the RRR ≈129 is much higher. The typical film thickness was ~0.4 µm for both depositions. Typical RRR for Nb cavity material is ~300, so the thin-film value of 129 is an encouraging result. Over a range of deposition conditions, it was observed that the RRR increased with film thickness up to about 2µm thickness, then leveled off. Thus the measured value might be higher still in a thicker film.

XRD of Thin-films

Figure 5 shows the XRD spectrum measured on Nb films coated on sapphire at 300°C. The x-ray peaks from bare sapphire (Al_2O_3) are also shown. The scans

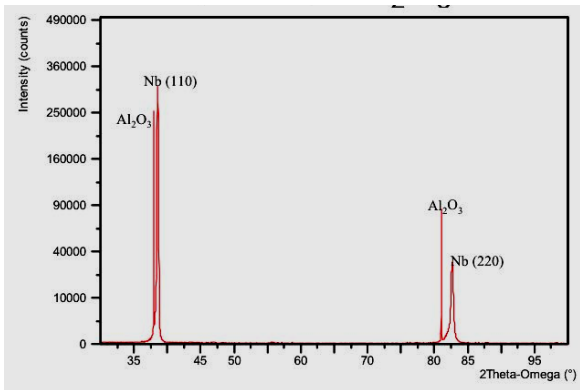

Figure 5: XRD spectrum of Nb on a-plane sapphire.

examined the orientation of planes that are parallel to the substrate surface, as well as any microstructural differences such as lattice constant and strain, between the Nb films and sapphire substrates. All of the intense x-ray diffraction peaks could be systematically indexed as the body center cubic (bcc) phase of niobium. No other impurity phase was observed in the x-ray study.

Figure 6 shows an expanded region of this spectrum, and also includes spectra from films coated at room temperature, at 300°C and at 400°C.

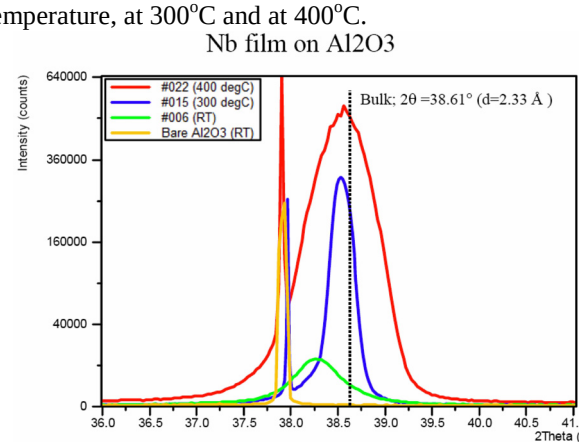

Figure 6: Expanded XRD spectrum, to reveal shape of peaks from films coated at different temperatures.

The dashed line in the figure shows the expected location of the peak for pure, bulk Nb (2θ=38.61°). As the substrate temperature is increased from room temperature, the location of the peak shifts closer to the bulk Nb location, suggesting that the 400°C coated film has almost bulk like structure, as least as indicated by the XRD spectrum. The broadening of the peak as one goes from 300°C to 400°C might be related to crystal grain orientation and not to structural defects per se. The peak is significantly broadened and also shifted as compared to the bulk niobium peak. Both the lattice parameter and the grain size may be estimated from this (110) peak. The former was calculated using Bragg's law, whereas the latter was found using the Debye-Scherrer formula [11]. A significant increase in the lattice parameter is observed in comparison with the bulk value (a=3.30Å). Such an increase in lattice parameter with decrease in grain size has been reported earlier for very thick films [12].

EBSD of Nb Thin-films

Figure 7 shows data obtained from EBSD on the Nb film that was coated at 300°C.

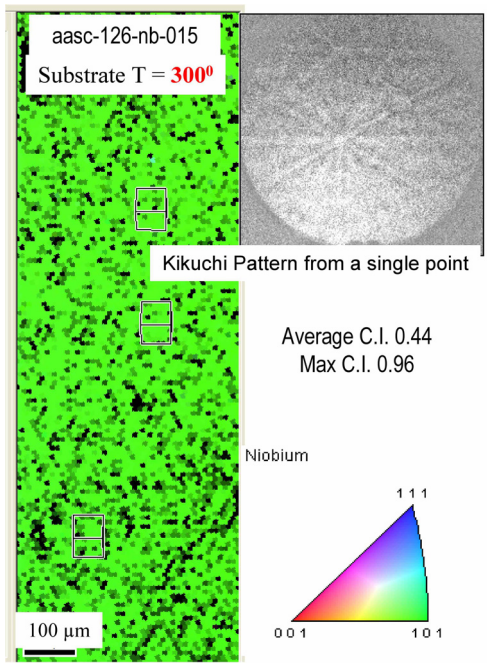

Figure 7: EBSD images from Nb film coated at 300°C.

The left portion shows the EBSD image with a 100µm bar for scale. The black dots indicate regions where the C.I. index was below an acceptance threshold. The triangle at right shows the color code for the crystal orientation. The fairly uniform green color across the entire field of view indicates a single orientation [110]. The Kikuchi pattern at top right (taken from a single spot) is sharp, consistent with the uniform orientation and the narrow XRD peak (blue curve) in Fig. 6.

Figure 8 shows similar data obtained from a film coated at 400°C. In addition to the EBSD data, Fig. 8 also shows an SEM image (left) at the same magnification (see 200µm bar) as was used for the EBSD image. Here we observe that the crystal orientation [110] is uniform over a 1mm x 0.6mm region on the film. The film thickness was only ≈0.4µm. Again a sharp Kikuchi pattern is observed. The SEM and EBSD images show typical crystal orientation cubes for reference.

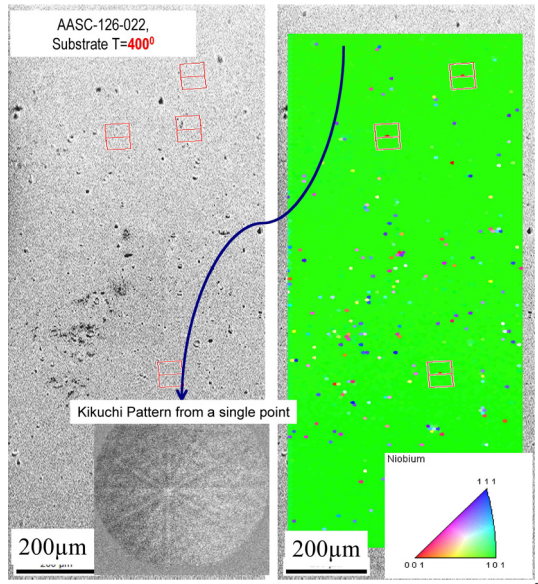

Figure 8: EBSD images from Nb film coated at 400°C.

COATING OF RF CAVITY STRUCTURES

A 2.2 GHz copper RF cavity half-cell was coated to demonstrate the feasibility of utilizing the CED™ process for this purpose. Figure 9 shows the coated half cell. No analysis was performed on the cavity to characterize the film. One observes that no un-coated regions or macroscopic defects are visible and the film appears well-adhered.

Figure 9: 2.2 GHz Copper RF cavity half-cell coated with Nb using the CED™ process.

Along with Superconductivity characteristics such as T_c and RRR, there are mechanical characteristics necessary to produce a functional SRF cavity from Nb coated Cu. Notably the film surface must be very smooth, with RMS roughness below approximately 150nm. SRF cavities drive accelerating gradients as large as 200 MV/m. At such large electric fields, electron generation at the surface of the cavity is a formidable problem. Electrons

produced at the cavity surface can scatter the accelerated beam, or result in multipacting, where electrons impact the cavity surface and release more electrons, which in-turn can cause beam scattering or still more multipacting. Field emission and multipacting cause heating of the cavity and can lead to a cryogenic quench. Thus roughness must be minimized. Cathodic arc deposition methods such as CED™ produce macro-particles which are μm scale droplets of material produced from the cathode surface. Cathode material leaves the cathode spot as a mixture of plasma and macro-particles. The ratio of plasma to macro-particles is a strong function of the thermodynamic properties of the cathode and the local current density. In the 1970's Daalder studied many cathode materials ranging from low melting point metals to refractory metals. The basic trend was that the fraction of the mass leaving the cathode as plasma increased with increased melting point of the cathode material. Daalder showed that the relative rate of macro-particle production (expressed as mass/coulomb) can be significantly reduced by reducing arc current. However it does so at the expense of deposition rate as less plasma is produced at lower arc currents. Thus a balance must be established between the large deposition rates desired to maintain low relative flux of impurities at the substrate surface and the low arc currents desired to reduce macro-particle production.

Likewise depositions can be performed at varying levels of applied axial B-field. It has been shown that increasing the cathode spot velocity causes a decrease in the production of macro-particles. Cathodic arc-spot velocity increases with increasing axial B-field.

DISCUSSION

Superconducting RF cavities enable research particle accelerators to achieve higher beam energies at lower operating powers. Superconducting cavities are made from niobium which is expensive and difficult to manufacture. Coating copper cavities with a thin film of Nb would save on both fabrication and operation costs and allows enhanced accelerator operation. This paper has described the use of energetic condensation using our CED™ process, to deposit thin-films of Nb on sapphire and Cu samples. The sapphire coated films allowed measurements of sharp superconducting transitions and RRR as a function of film thickness and substrate coating temperature. A dramatic increase in RRR with substrate temperature, from ~5 at room temperature to 50 at 300°C and 129 at 400°C was measured. The RRR was also observed to increase with film thickness up to 2μm. Hence the measured value of 129 for a 0.5μm film is a lower bound. These measurements were complemented by measurements of surface properties using SEM, XRD and EBSD. EBSD images showed uniform crystal orientation [110] over fairly large fields of view (~1mm x 0.5mm), across the 0.5μm thick Nb film. XRD spectra showed that the thin-film Nb crystal peaks approached the 2θ angular position of bulk Nb as the substrate temperature was increased to 400°C.

In general, there is a complex interplay between vacuum pre-bake conditions and deposition conditions that determines grain structure, film thickness and good superconductivity in such Nb thin-films. Future work will further study the morphology of the film using SEM, EBSD, XRD and other tools, optimize the pre-bake and deposition conditions, followed by RF tests in a superconducting impedance cavity (SIC) at Jlab.

ACKNOWLEDGMENTS

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