Diffusion of Nitrogen into Niobium: A Surface Treatment for RF Cavities

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This paper discusses the effects of doping superconducting radio frequency (RF) cavities. A series of cavities prepared at Cornell were baked in a low pressure atmosphere of nitrogen following heat treatment in vacuum. The results show a boosted medium field quality factor and quench fields as high as 32 MV/m. The data supports reports from FNAL and JLAB on the benefits of nitrogen doping. Investigation of nitridation kinetics and surface analysis has provided new insights concerning the mechanism behind the treatment.

Introduction

Superconducting RF cavities accelerate charged particles which can be used to study high energy particle physics and generate high luminosity light sources. The quality of these cavities and their ability to perform at high energies can be a limiting factor in the design of particle accelerators. Nitrogen treatment is a relatively new technique that has been shown to improve the quality factor of niobium cavities. The cavities are baked in a low pressure nitrogen atmosphere and the gas is allowed to react and diffuse into the metal. Chemical analysis is providing clues about the material properties and how the treatment works. Constructing more accurate models will hopefully lead to developing cavities that fit the specifications needed for future accelerators such as Stanford's LCLS-II project and Cornell's ERL project.

Cavity Treatment and Vertical Testing

The information presented in this paper concerns a collection of five single-cell 1.3 GHz ILC shaped cavities as well as three small niobium disks which received the same treatment in a low-pressure nitrogen atmosphere. All of the samples were prepared at Cornell from RRR 300 niobium. To begin with, the cavities underwent 100 μm of vertical electropolishing (EP). The cavities were then baked in vacuum for 4 days at 800°C. As the vacuum heat treatment drew to a close, nitrogen gas was introduced to the furnace for a duration of 20 minutes. Initially, a pressure of 38 mTorr was injected. As nitrogen absorption proceeded, the pressure fell to 30 mTorr at which point more gas was injected to reach a maximum pressure of 57 mTorr. The diffusion of the gas into the metal was allowed to continue uninterrupted until the 20 minutes were over. The chamber was then pumped back to vacuum for an additional 30 minutes of backing. After the heat treatment another round of EP was administered. Each cavity received at least 5 μm of material removal. The cavities were all tested at Cornell in vertical testing dewars. Measurements were taken of the quality factor (Q_0) and the resonance frequency. The dependence of these characteristics on temperature and applied accelerating field was evident in the data. Graphs were fitted by programs such as SRIMP which solves BCS theory to extract material properties. The critical temperature, mean free path, energy gap, and residual resistance associated with each cavity were of particular interest. Surface analysis tools such as Secondary Ion Mass Spectrometry (SIMS) and X-ray Photoelectron Spectroscopy (XPS) were used to quantitatively investigate the nitrogen content of the niobium disks.

Cavity Performance

The results of vertical testing on the last nitrogen-doped cavity (TE1-5) in the series are shown in Figure 1. Electromagnetic waves are fed into the cavity. By fine tuning the frequency around 1.3 GHz, stable standing waves can be contained. To test the quality factor, experimenters shut off the RF power and measure the time it takes for the field to decay. The amount of decay time is directly related to the amount of dissipated power which allows Q_0 to be calculated. LCLS-II specifications require a Q_0 of at least 2.7 x 10^{10} at 2.0 K and 16 MV/m. An untreated cavity provided a baseline Q_0 of approximately 1 x 10^{10} at 2.0 K and 16 MV/m. It can be seen that the doped TE1-5 cavity meets the requirement with a Q_0 of 3 x 10^{10} at 2.0 K.

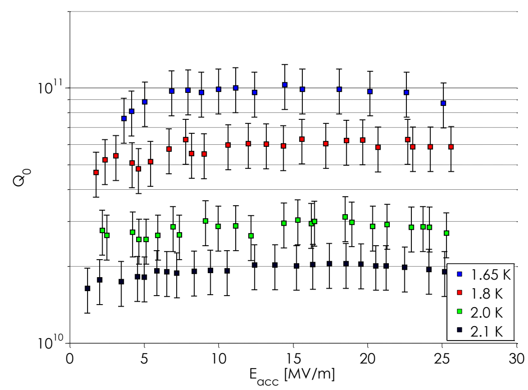

Figure 1: Q_0 vs E_{acc} at different temperatures for TE1-5.

The other four cavities in the collection also exceed the specifications with an average Q_0 of 4 x 10^{10} at 2.0 K and 16 MV/m. The undesirable degradation of quality at fields above 16 MV/m (medium field Q_0-slope) is a limitation that has plagued niobium cavities. Superconducting cavities dissipate an unbelievably small fraction of the total applied energy, but they still experience surface resistance. This resistance leads to dissipation and a lower Q_0. Surface resistance consists of BCS resistance (which is dependent on temperature and applied field) and residual resistance (which is caused by impurities in the surface layer). As greater fields are applied, the BCS resistance has an increasingly negative effect on the cavity performance. Doping appears to counteract this effect. Introducing nitrogen to niobium decreases the mean free path and increases the energy gap. A decrease in BCS resistance is also observed from standard niobium while the residual resistance is comparable between doped and standard. For this reason, it is believed that changes in BCS material properties are the driving force behind the Q_0 improvement.

Surface Analysis

The chemical composition of the cavities was not tested directly, but related to the chemical composition of the niobium disks which were treated using all of the same material, equipment, and procedures. Depth profiles were measured to provide information about the nitrogen diffusion. XPS analysis revealed cursory information about the concentration of nitrogen in the first 2 µm of the material (Figure 2). The existing models for nitridation kinetics predicted that a layer of niobium nitride (NbN) should form at the surface of the sample. The concentration of nitrogen in this phase was expected to be approximately 50%, but a maximum of 30% was measured. Preferential sputtering of nitrogen may contribute to this low relative abundance, but it does not explain the 20% discrepancy.

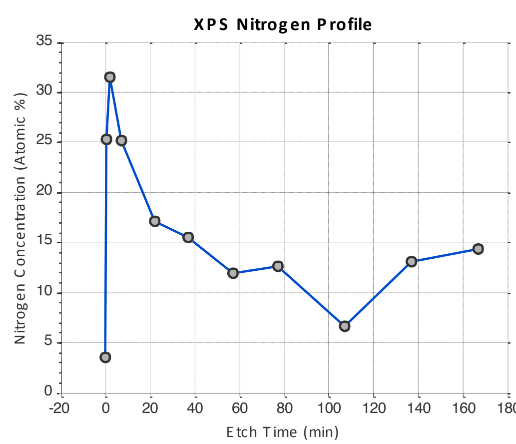

Figure 2: A profile of nitrogen concentration in the surface of a niobium sample (depth is less than 2 µm).

SIMS data (Figure 3) more clearly illustrates the location of the nitride layer as well as the phase following the nitride transition. This inner phase incorporates nitrogen as an interstitial, occupying the space between niobium atoms in the metal lattice.

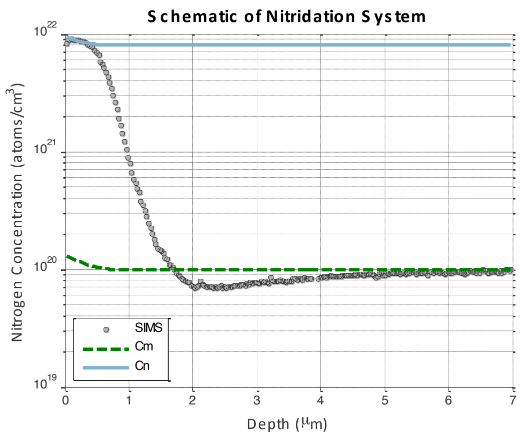

Figure 3: Raw data of a depth profile with overlaid models for the concentration of nitrogen in the nitride layer (Cn) and in the metal (Cm).

The portion of the SIMS curve corresponding to NbN also displays a lower than expected nitrogen level. It may be that the nitride conversion takes place predominantly along grain boundaries in the niobium. Niobium can have grains that are micron scale. As we probed further into the sample, we may have cut through the boundaries and exposed metal that had not been converted into NbN. This would average and dilute the concentration signal. To reach peak performance, the cavities receive EP to remove the nitride layer. The non-uniformity caused by grain boundaries may also explain the low Q_0 associated with this phase. The dip in nitrogen concentration following the transition (2-3 μm) remains a mystery. Further investigation is currently being conducted to generate a depth profile 100 μm into the sample. This should be deep enough to determine if the nitrogen concentration has leveled off. Varying treatment parameters such as nitrogen pressure, baking temperature and material removal will hopefully increase understanding of diffusion kinetics. Eventually, the treatment may reveal an optimum ratio of nitrogen to niobium as well as a procedure to create an even phase with the "optimum dirtiness."

Conclusion

Treatment in a low-pressure nitrogen atmosphere has been shown repeatedly to combat mid-field Q_0 slope and yield cavities that meet specifications for future particle accelerators. Perhaps acting as an interstitial, nitrogen is able to decrease the mean free path and increase the energy gap. This decreases the temperature dependent BCS resistance and leads to a cavity whose performance does not diminish at fields of 16 MV/m. The consistently high quality of nitrogen-doped cavities with only rudimentary understanding of treatment parameters is a testament to the potential of doping.

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