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Experimental evaluation of niobium film pinholes

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Abstract

To assess the quality of vacuum-deposited films, well-defined areas of free-hanging thin film may be obtained by chemically dissolving the substrate on a fraction of its surface. A global evaluation of the thin film pinholes is then possible by measuring the gas conductance across the film. To do so, copper discs clamped between Con-Flat flanges have been used to separate a chamber filled with helium gas from another chamber where the helium throughput was measured. This method has been used to compare films produced under dust-free and dusty conditions, of various surface roughnesses and at different deposition incidence angles.

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1. INTRODUCTION

Depending on the requirements of a given application, many different techniques may be used to assess the quality of vacuum deposited thin films. Most of these techniques are adequate to determine average characteristics of coatings: electrical measurements may be used to obtain the resistivity, the purity may be determined by surface analysis, the crystal structure by X-ray spectroscopy, and so on.

Localised defects, such as pin-holes, are usually detected by scanning electron microscopy, a time consuming, tedious operation which furthermore does not provide average quantitative information. In the case of the superconducting RF cavities developed at CERN [1] and produced by industry [2] to upgrade the energy of the CERN Large Electron Positron (LEP) Collider, the superconducting function is provided by a thin film of niobium deposited on the inner walls of a copper cavity by magnetron sputtering. In this case pin-holes in the Nb film may be very detrimental to cavity performance, because they may expose the underlying copper, which is about 10^5 times more resistive to RF power than the superconducting Nb at 4.2 K. As a consequence, the cavity surface resistance would be higher and the cryogenic losses correspondingly increased. Considering the ratio of the Nb and Cu resistivities, the uncoated copper surface area should not exceed about 10^{-6} of the cavity surface.

Pin-holes may be produced in a thin film coating by various causes. Dust particles sitting on the surface may be embedded in the coating and leave a hole when breaking loose. Surface protrusions may result in uncoated areas by shadowing, particularly when the coating atoms impinge on the substrate at grazing angles. Substrate pitting, often produced by chemical surface etching, may replicate in the coating, depending on pit size and coating thickness. All these different causes have been investigated individually, making use of an innovative technique developed for this purpose.

This technique consists in measuring the gas throughput across a known area of bare film. This measurement allows the global conductance of the pin-holes in the coating to be measured, which is then normalised to the total bare film virtual conductance to obtain the unquoted area fraction. The procedure developed is described in detail below.

2. EXPERIMENTAL SET-UPS DESCRIPTION

The chosen substrates were oxygen-free copper disks having the same diameter and thickness as a DN40CF gasket, namely 48 mm and 2 mm respectively. They were thinned at the centre on a 1 cm diameter area down to a thickness of 0.3 mm by mechanically removing copper from just one side of the disks. Before coating the substrates were degreased and a 100 μm thick surface layer removed by electropolishing. Some of the substrates were chemically polished (about 10 μm removed) in an acid solution called SUBU5 (see Appendix 1) as routinely carried out for the LEP superconducting cavities.

The flat surface of the disks was finally coated with a 1.5 μm thick Nb film by the magnetron sputtering technique. During the coating process the discharge voltage was 400 V, the discharge current 2.5 A and the Ar pressure 8×10^{-4} mbar. The samples were fixed in the sputtering system in such a way as to allow a nearly perpendicular or a grazing angle (27°) of incidence of the Nb sputtered atoms. The coating process was performed under normal dusty conditions or following a dust-free process in a class 100 clean room.

A small fraction of the coated surface of the samples was then observed by scanning electron microscopy (SEM) to identify some of the major film defects such as pitting, lack of adherence or scratches.

To obtain a free-hanging Nb film in the thinned central area of the sample, the coated disks were dipped into an acid solution (see Appendix 2), which selectively attacked copper but not niobium. The acid attack lasted about 5 hours; after that time the 0.3 mm thickness of the central thinned Cu part was completely removed therefore leaving a bare unsupported Nb film surface of about 0.7 cm^2 .

The samples were then mounted on an optical microscope that allows the automatic detection of light spots transmitted through the pin-holes in the bare Nb film when illuminated from the back. Due to light diffraction only qualitative information can be obtained with this technique.

Finally the samples were mounted on the vacuum system that allowed the evaluation of the pin-hole surface area by measuring the He leak rate through the bare Nb film. The samples, which were clamped between two DN40CF flanges, divided the vacuum system in two parts (see Fig.1). Once a pressure in the system lower than 1×10^{-8} mbar was reached, the valve V1 was closed and He was injected up to 2×10^{-1} mbar through a variable leak valve in the high pressure side of the system. The pressure was monitored on the low pressure side and the He leak rate was then evaluated. The flux passing through the film is obtained by evaluating the turbopump throughput and then used to calculate the fraction of uncoated area.

Care must be taken when pumping down the system to prevent creating an excessive pressure difference across the bare film. This can be achieved by slowly opening the valve V2 mounted on the roughing pump while leaving the valve V1 completely open.

The lowest detectable uncoated fraction is given by the maximum pressure of gas that can be injected in the high pressure side and by the lowest recordable pressure in the low pressure side of the system. The first is limited by the weak mechanical strength of the unsupported film; maximum pressures of about 10^{-1} mbar are a reasonable limit. The second is dictated by the sensitivity of the measuring instruments; in a baked system pressures as low as 10^{-12} mbar can be detected by means of residual gas analysers providing a secondary electron multiplier is available. It results that an uncoated fraction down to values as little as 10^{-10} could be measured.

3. EXPERIMENTAL RESULTS AND DISCUSSION

As expected SEM pictures revealed the presence of uncoated zones and large defects on the Nb film grown under dusty conditions. Pitting on the underlying Cu substrate was shown only on samples chemically polished by SUBU5 (growth under both clean room and dusty conditions). The density of the pits was about 500 mm^{-2} and their diameter was in the range of $1 \text{ }\mu\text{m}$. SEM pictures seem to indicate that these pits on the copper substrate are completely covered by the Nb film so that they should not be a source of pin-holes.

No transmitted light spots have been detected by optical microscopy in clean room grown films. On the other hand spots as large as $9000 \text{ }\mu\text{m}^2$ were identified in films grown under dusty conditions. The distribution of the spot size did not follow any rule indicating that the dust particle collection on the substrate surface before coating is completely accidental. Nevertheless the smallest spot size detected on samples grown with normal incidence was always smaller than that found on samples grown with grazing incidence. This could be due to the fact that the shadow of the dust particles is enlarged when the Nb atoms impinge on the surface with a small angle.

The values of the uncoated area fraction measured by the He flux rate method are reported in Tab.1. The films grown under clean room conditions have a pin-hole fraction of about 10^{-7} while this value is at least two orders of magnitude larger for film grown under dusty conditions. As indicated by the SEM pictures no significant differences have been recorded between samples treated with or without SUBU5 but enlarged statistics are needed to substantiate this preliminary result.

Samples coated with a small incidence angle have shown, compared to those deposited under normal incidence, an unexpected lower He flux rate. This could be explained by a different dust particle collection in the sputtering system due to the different point of mounting, the samples coated with a small angle of incidence were face down while those coated under normal incidence were positioned vertically.

4. CONCLUSIONS

A new method for the measurement of pin-hole area fraction in thin films has been developed. It consists of measuring the He flux rate through a defined area of unsupported film. The lowest limit of detection of the uncoated fraction can be extremely low and has been estimated to be of the order of 10^{-10} . This limit is surely lower than that obtained with other techniques, such as electrolytic current measurements [3], due to the absence of signal noise.

Values of uncoated fraction of about 10^{-7} were obtained for Nb films deposited on copper under clean room conditions. If the deposition is performed under dusty conditions, this fraction is at least two orders of magnitude larger.

The number of measured samples is at present too small to settle any argument concerning the effect of the incidence angle or of the acid polishing. Furthermore, the very smooth surface produced by the electropolishing and not dramatically altered by the subsequent slight chemical polishing may contribute to reduce the difference between the differently treated samples. Larger differences may be found when comparing electropolished samples to samples chemically treated only, provided however that a reasonable statistics is obtained. This point deserves a further investigation. Other combinations of coating and substrate materials will be also examined in view of the growing CERN interest for non-evaporable getter (NEG) coatings.

REFERENCES

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APPENDIX 1

The SUBU5 chemical polishing solution consists of:

- sulfamic acid 5 g l⁻¹;
- hydrogen peroxide 5% vol;
- n-butanol 5 % in vol;
- ammonium citrate 1 g l⁻¹.

The treatment is carried out at 72°C, preceded and followed by washing with sulfamic acid.

APPENDIX 2

The acid solution used to remove the copper substrate consists of:

- chromic acid 80 g l⁻¹;
- sulfuric acid 3 ml l⁻¹;
- demineralised water.

The treatment is performed at room temperature.

Table 1 - Values of the uncoated fraction of Nb film coated in clean and dusty condition on copper disks treated in two different ways. Symbols $\perp$ and $\angle$ indicate normal and grazing angle incidence of the sputtered atoms on the substrate respectively

ENVIRONMENT	SURFACE TREATMENT	UNCOATED FRACTION
DUSTY CONDITIONS	Electropolishing	$\perp$ 1.2×10^{-4} $\angle$ 9.4×10^{-5}
	Electropolishing + SUBU5	$\perp$ 1.5×10^{-4} $\angle$ 3.2×10^{-5}
CLEAN ROOM	Electropolishing	$\perp$ 1.9×10^{-7}
	Electropolishing + SUBU5	$\perp$ 3.5×10^{-7}

Figure 1 - Schematic view of the vacuum system used for the helium leak flux rate measurements through the unsupported film.